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Rhodium-catalyzed intermolecular C (sp3)-H amination in a purely aqueous system

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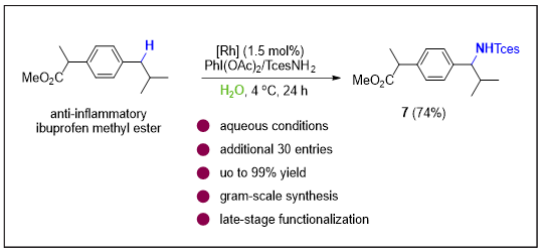

In recent years the synthetic community has been experiencing an increasing pressure and motivation on the way to improve the environmental benignity of chemical processes. However, despite considerable advances there remains a significant challenge for synthetic systems to perform transformations that nature can routinely achieve at physiological conditions. Considering the green nature of water, the aqueous medium adopted in enzymatic processes is of immense interest to the synthetic community. However, reactions mediated by synthetic catalysts, even for most prominent transition metal complexes that have revolutionized industrial organic synthesis are still primarily restricted to organic solvents and have met with limited success when used in water. Aliphatic amines represent a category of compounds particularly favored by nature as validated by the wide occurrence of diverse biologically active alkaloids. Conventional approaches mainly rely on the inherent reactivity of functional groups such as alcohol and carbonyl, the direct C-H amination via transition metal catalyzed nitrene transfer provides a novel and straight forward synthetic strategy. Given the high electrophilicity of metal nitrenoid intermediates, such amination processes are normally performed in inert solvents not bearing C (sp^3)-H bonds (e.g. benzene, dichloromethane). In this context, we have successfully developed the first metal catalyzed intermolecular C (sp^3)-H amination reactions performed in a purely aqueous medium under mild conditions. The method features great environmental benignity and high efficiency towards a wide range of hydrocarbons bearing different functional groups. Its versatile synthetic utility has been demonstrated by late stage functionalization of several bioactive molecules and thus we anticipate the methodology described here will find wide applications in sustainable chemistry of catalytic C-H functionalization.

Recent Publications

1. X Lu, Y Shi and F Zhong (2018) Rhodium-catalyzed intermolecular C (sp3)-H amination in a purely aqueous system. *Green Chemistry*; 20: 113.
2. T Wang, X Han, F Zhong, W Yao and Y Lu (2016) Amino acid-derived bifunctional phosphines for enantioselective transformations. *Accounts of Chemical Research*; 49(7): 1369-78

Biography

Fangrui Zhong has obtained his BSc from Zhejiang University followed by his PhD. He had joined the Faculty of Huazhong University of Science and Technology as a Professor of Chemistry. He has received several awards including Thieme Chemistry Journal Award; Singapore National Institute of Chemistry Gold Medal; Wang Gungwu Medal and Prize and finalist of Reaxys PhD prize. His research involves the development of novel and efficient sustainable synthetic methods inspired by nature.

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